

Estimation of Critical Constants T_c , ρ_c from the $\rho(T)$ and $T(\rho)$ Relations at Coexistence*

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The critical temperature T_c and the critical density ρ_c are estimated from new equations (1) for the double-valued $\rho(T)$ relation, and (2) for the single-valued $T(\rho)$ relation. For method (1) this report gives concise descriptions of the rectilinear diameter and of the densities of liquid oxygen. Calculated vapor densities agree well with experimental oxygen data over the wide range from triple- to critical-point. Method (2) is applied to hydrogen, oxygen, fluorine, and neon, giving the $T(\rho)$ relation and estimates for T_c and ρ_c all in one step from the complete set of liquid and vapor data.

Key words: Coexistence; coexistence temperatures; critical constants; critical density; critical temperatures; fluorine; neon; oxygen; orthobaric densities; parahydrogen; saturated liquid and vapor densities.

List of Symbols

Subscripts

c refers to the critical point.
 t refers to the liquid triple point.
 g refers to vapor at coexistence.
 l refers to liquid at coexistence.

Symbols

a, b, c, d, e, f, g , constant coefficients.
 A_i , constant coefficients.
 β , an exponent near 0.35.
 d , density at coexistence.
 d_t , 38.203 mol/l for parahydrogen [8].
 d_t , 40.83 mol/l for oxygen [4].
 d_t , 45.2 mol/l for fluorine (estimated).
 d_t , 61.785 mol/l for neon [23].
 l , the liter.
 P , pressure.
 $\rho \equiv d/d_t$, density reduced at the liquid triple point.
 $\sigma \equiv d/d_c$, density reduced at the critical point.
 T, K , temperature at coexistence (NBS 1955).
 $\tau \equiv T/T_c$, temperature reduced at the critical point.
 $z \equiv (1 - \tau)$, reduced temperature with origin at T_c .

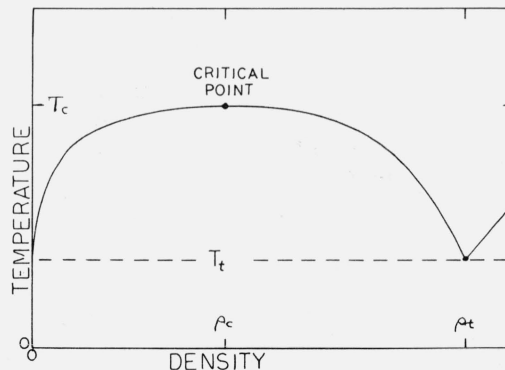


FIGURE 1. Temperature-density phase diagram for coexisting vapor and liquid at densities to the liquid triple point, ρ_t .

1. Introduction

This report completes protracted efforts to forge some tools needed for the computation of thermodynamic properties of fluids from experimental PVT data. These tools are concise descriptions of the data, descriptions which are essentially correct in the critical region where PVT data are least reliable. For the one-phase domain we may use the nonanalytic equation of state [1].¹ For the two-phase domain we may use the nonanalytic vapor pressure equation [2] together with the $\rho(T)$ or $T(\rho)$ relations described below and outlined by figure 1.

¹ Figures in brackets indicate the literature references at the end of this paper.

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2. The Orthobaric Densities

Guggenheim [3] has formulated approximate, general relations for the double-valued function $\rho(T)$. (Symbols are listed above.)

$$(\rho_l/\rho_c - 1) = (3/4) \cdot z + (7/4) \cdot z^{1/3}, \quad (1a)$$

$$(\rho_g/\rho_c - 1) = (3/4) \cdot z - (7/4) \cdot z^{1/3}. \quad (1b)$$

With precise data for oxygen [4] we find that (1a, 1b) are valid only in the approximate range $0.85 < T/T_c \leq 1$. Hou and Martin [5] extended the range for liquid densities by use of an expansion in powers of $z^{1/3}$.

Equations (1a, 1b) are consistent with the hypothesis of a rectilinear diameter,

$$(\rho_l + \rho_g)/2 = \rho_c + a \cdot z. \quad (1c)$$

They also are consistent with the currently accepted relation for the critical region

$$(\rho_l - \rho_g)/2 = b \cdot z^\beta \quad (1d)$$

where exponent $\beta = 0.350 \pm 0.015$ [6]. Again we find with oxygen data that neither (1c) nor (1d) is sufficient at $T/T_c < 0.9$. Oxygen vapor densities range through a factor greater than 60,000 [4]. To secure a precision of at least 0.03 percent in calculated vapor densities for thermal computations near the triple point, we would require a precision of about one part in 400 million in each of two functions to be differenced as in (1b).

In the following we give results of investigations on the $\rho(T)$ relations, using oxygen data from [4]. The method is awkward. Raw data for $\rho_g(T)$ and for $\rho_l(T)$ must be interpolated to the same temperatures before they can be combined.

The critical temperature is obtained by trial to minimize deviations in the logarithmic form of (1d). The coefficients $\log(b)$ and β are found by least squares. For oxygen data this expression is sufficient only in the restricted range $0.94 < T/T_c \leq 1$. Summary results are in table 1.

The critical density is obtained as the value of the rectilinear diameter (1c) at $T = T_c$. The data unfortunately are not linear, but rather are slightly sigmoid, as described within the high precision of these data by the new expression,

$$(\rho_l + \rho_g)/2 = a + b \cdot z + c \cdot \exp(-d/z) \quad (2)$$

in which $\rho_c = a$. Equation (2) is consistent with the hypothesis of a rectilinear diameter because it becomes exactly linear as $T \rightarrow T_c$. Summary results are in table 1.

The densities of liquid oxygen have been examined for deviations from forms like (1a). We thus deduce the new expression ($\beta = 0.35$),

$$(\rho_l/\rho_c - 1) = a \cdot z + b \cdot z^\beta + c \cdot \exp(-d \cdot \tau^2/z). \quad (3)$$

Equation (3) is consistent with (1c) and (1d) because the last term vanishes strongly as $T \rightarrow T_c$. The values for a and b are close to those generalized by Guggenheim, and the last term is merely a small adjustment, as seen by the value of c in table 1.

The densities of oxygen vapor are described quite well by the following expression,

$$\rho_g/\rho_c = \exp[a \cdot (1 - 1/\tau) + b \cdot z^\beta + c \cdot z + \dots + g \cdot z^5]. \quad (4)$$

It is consistent at low densities with the ideal gas law and the basic vapor pressure equation. This exponential form excludes negative values for ρ_g , as often are obtained with difference expressions like (1b). Exponents other than $\beta = 0.35$ have not been explored. Summary results are in table 1.

TABLE 1. Summary results for oxygen with equations of section 2

Equation (1d).	10 points at $145 \leq T \leq 154$ K,	$T_c = 154.54$ K,
	$b = 24.618$ mol/l,	$\beta = 0.3476$, rms = 0.35%.

Following equations for 53 points at $56 \leq T \leq 150$ K

Equation (2).	$a = 13.6135$ mol/l,	$b = 9.7843$ mol/l,
	$c = 1.9306$ mol/l,	$d = 0.9195$, rms = 0.020%.
Equation (3).	$a = 0.7441$,	$b = 1.8212$, $c = -0.0556$,
	$d = 5/7$, rms = 0.037%.	
Equation (4).	$a = 3.2305$,	$b = -1.9368$, $c = -1.6989$,
	$d = 4.7351$, $e = -24.2531$, $f = 45.7606$,	
	$g = -46.7552$, rms = 0.030%.	

3. The Saturation Temperatures

The $T(\rho)$ relation for coexisting (saturated) liquid and vapor is single-valued (fig. 1) and therefore requires no interpolation of data for analytical work. As it is the heart of the nonanalytic equation of state [1], we have sought a description which would be simpler and more accurate than reported [7]. With symbols defined in the list, the result of new investigations with parahydrogen data of [8] is the following,

$$(1/\tau) - 1 = |\sigma - 1|^3 \cdot F(\rho) \quad (5)$$

where

$$F(\rho) \equiv A_1 \cdot \log_e(1/\rho) + A_2 + A_3 \cdot \rho + \dots + A_9 \cdot \rho^7.$$

Equation (5) is consistent with the ideal gas law and the basic vapor pressure equation at low densities. In the critical region it is consistent with the well-known [3] behavior, $\tau = 1 - A \cdot |\sigma - 1|^3$. At high densities we obtain $\tau \rightarrow 0$ (because $A_9 > 0$), consistent with the trend of figure 1. The argument ρ is selected because it has a maximum value of unity: we therefore may keep the same number of decimal digits in all coefficients A_i . For least squares, (5) has been used exactly as written. When the number of terms is increased from eight to nine, the rms relative deviation drops by

a factor of 1/6 for the hydrogen data [8]. Nine terms also is optimum for the oxygen data [4].

In the following we estimate the critical constants via (5) for hydrogen, oxygen, fluorine, and neon. Simultaneously we obtain a complete description of the $T(\rho)$ relation for vapor and liquid in a single formula, as compared with at least four formulas required by the familiar but awkward methods of section 2 above. Some data near ρ_c will be omitted arbitrarily from the calculations (see tables 3 and 5) because they may be of questionable accuracy, having been obtained from PVT data and a vapor pressure equation [4, 8].

We vary the assumed values for T_c and for ρ_c in (5), observing the root mean square (rms) of relative deviations ($T/T_{\text{calc}} - 1$). First results for hydrogen are shown by figure 2. Similar results at a higher precision are in table 2. For each density ρ_c the first column gives T_c and the second column gives the relative rms deviation in parts per ten thousand. The minimum in the deviation is quite well defined. Table 3 compares individual reduced data with calculated values. Table 6 gives the coefficients for (5). Table 7 compares critical constants estimated via (5) with estimates of other authors.

TABLE 2. Parahydrogen survey for T_c , ρ_c in eq (5)

T_c , K	rms $\cdot 10^4$	T_c , K	rms $\cdot 10^4$
DCRT, mol/l = 15.550		DCRT, mol/l = 15.610	
32.944	1.35	32.944	1.18
32.947	1.21	32.947	0.99
32.950	1.12	32.950	.86
32.953	1.10	32.953	.79
32.956	1.14	32.956	.82
32.959	1.24	32.959	.93
32.962	1.39	32.962	1.10
DCRT, mol/l = 15.570		DCRT, mol/l = 15.630	
32.944	1.21	32.944	1.29
32.947	1.05	32.947	1.12
32.950	0.93	32.950	1.00
32.953	.89	32.953	0.94
32.956	.93	32.956	.95
32.959	1.04	32.959	1.04
32.962	1.21	32.962	1.19
DCRT, mol/l = 15.590		DCRT, mol/l = 15.650	
32.944	1.15	32.944	1.48
32.947	0.97	32.947	1.32
32.950	.83	32.950	1.21
32.953	.78	32.953	1.16
32.956	.81	32.956	1.17
32.959	.93	32.959	1.24
32.962	1.11	32.962	1.36

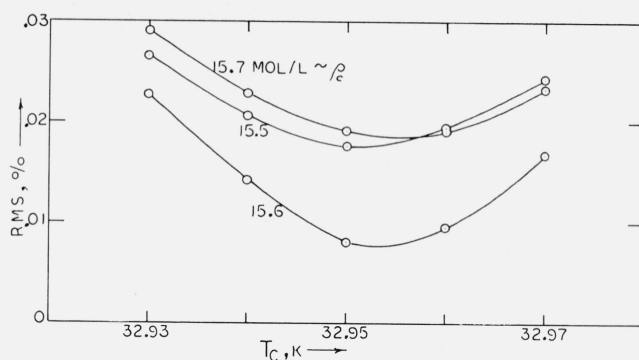


FIGURE 2. Relative rms deviation of hydrogen temperatures from eq (5) as a function of assumed T_c , ρ_c values.

The oxygen survey in table 4 is analogous to that for hydrogen. Whereas the minimum in the overall deviation is weak (as compared with that for H_2), the data extend over a much greater range of vapor densities. Table 5 compares individual data with calculated values of T/T_c . Table 7 compares critical constants.

Fluorine PVT measurements are in progress [16], and accurate vapor pressures have been reported [17]. We have obtained orthobaric densities via the vapor pressure relation and an equation of state (similar to that in [14]), kindly provided by Prydz and Straty [16].

TABLE 3. Comparison of $T(\rho)$ data for parahydrogen with calculated values from (5). Part I, vapor densities

$d_c = 15.59 \text{ mol/l}, T_c = 32.953 \text{ K}$			
d/d_c	T/T_c	Calc	Pent
0.0641	0.6642	0.6642	0.00
.0962	.7180	.7180	-.00
.1283	.7592	.7592	-.00
.1604	.7926	.7926	.00
.1924	.8206	.8206	.00
.2245	.8445	.8445	.00
.2566	.8651	.8652	-.00
.2886	.8832	.8832	-.00
.3207	.8990	.8991	-.00
.3528	.9130	.9130	-.00
.3849	.9254	.9254	.00
.4169	.9364	.9363	.00
.4490	.9461	.9460	.01
.4811	.9547	.9546	.01
.5131	.9622	.9622	.00
.5452	.9689	.9689	.00
.5773	.9747	.9747	-.01
.6094	.9798	.9798	-.01
.6414	.9841	.9842	-.01
.6735	.9879	.9880	-.01
.7056	.9910	.9911	-.01
.7377	.9936	.9936	-.00
.7697	.9957	.9957	.00
.8018	.9974	.9972	.02

TABLE 3, PART II. Parahydrogen liquid densities

d/d_c	T/T_c	Calc	Pcnt
1.1867	0.9980	0.9978	0.02
1.2187	.9966	.9965	.01
1.2508	.9948	.9947	.00
1.2829	.9925	.9926	-.00
1.3149	.9899	.9900	-.01
1.3470	.9868	.9868	-.01
1.3791	.9831	.9832	-.01
1.4112	.9790	.9790	-.00
1.4432	.9743	.9743	-.00
1.4753	.9691	.9690	.00
1.5074	.9633	.9632	.01
1.5394	.9569	.9568	.01
1.5715	.9499	.9498	.01
1.6036	.9422	.9422	.01
1.6357	.9340	.9339	.01
1.6677	.9251	.9250	.00
1.6998	.9155	.9155	-.00
1.7319	.9052	.9052	-.00
1.7640	.8942	.8943	-.01
1.7960	.8825	.8825	-.01
1.8281	.8700	.8700	-.01
1.8602	.8567	.8567	-.01
1.8922	.8425	.8426	-.01
1.9243	.8276	.8276	-.00
1.9564	.8117	.8117	.00
1.9885	.7949	.7948	.01
2.0205	.7771	.7770	.01
2.0526	.7583	.7582	.01
2.0847	.7384	.7384	.01
2.1167	.7175	.7174	.00
2.1488	.6953	.6953	-.01
2.1809	.6719	.6720	-.01
2.2130	.6472	.6473	-.01
2.2450	.6212	.6212	-.01
2.2771	.5937	.5936	.02
2.4505	.4189	.4189	-.00

Summary results for (5) on saturated liquid and vapor from 86 through 142 K are given in table 6. Estimates for T_c and ρ_c are made only to ± 0.05 in each value because the data are preliminary. Table 7 compares critical constants.

Neon has been investigated with (5) to extend the range of types of substance. No determination of the critical density appears to have been made since the pioneer work of Mathias et al. [21]. To their orthobaric densities we have added the recent data of Gibbons [22]. Temperatures in [21] are adjusted to the ice point 273.15 K as in [23]. The combined data yield 16 vapor densities from 25.16 through 44.31 K, and 15 liquid densities from 25.16 through 44.08 K. Deviations from (5) are plotted in figure 3. Summary results are in table 6. The estimate for T_c is made only to ± 0.01 K; that for ρ_c only to ± 0.05 mol/l. Table 7 compares critical constants.

TABLE 4. Oxygen survey for T_c , ρ_c in eq (5)

T_c , K	RMS $\cdot 10^4$	T_c , K	RMS $\cdot 10^4$
DCRT, mol/l = 13.480		DCRT, mol/l = 13.540	
154.460	5.46	154.460	5.43
154.480	5.44	154.480	5.42
154.500	5.44	154.500	5.41
154.520	5.43	154.520	5.41
154.540	5.44	154.540	5.42
154.560	5.45	154.560	5.43
154.580	5.47	154.580	5.45
DCRT, mol/l = 13.500		DCRT, mol/l = 13.560	
154.460	5.44	154.460	5.44
154.480	5.43	154.480	5.43
154.500	5.42	154.500	5.42
154.520	5.42	154.520	5.42
154.540	5.42	154.540	5.43
154.560	5.43	154.560	5.45
154.580	5.45	154.580	5.47
DCRT, mol/l = 13.520		DCRT, mol/l = 13.580	
154.460	5.43	154.460	5.46
154.480	5.42	154.480	5.45
154.500	5.41	154.500	5.44
154.520	5.41	154.520	5.44
154.540	5.42	154.540	5.45
154.560	5.43	154.560	5.47
154.580	5.45	154.580	5.49

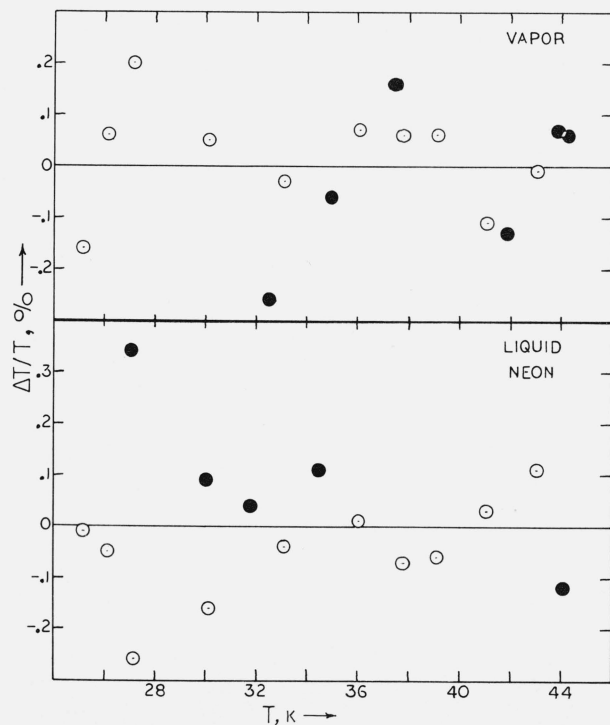


FIGURE 3. Relative deviations of neon saturation temperatures from eq (5).

TABLE 5. Comparison of $T(\rho)$ data for oxygen with calculated values from (5). Part I, vapor densities

$d_c = 13.52 \text{ mol/l}, \quad T_c = 154.52 \text{ K}$			
d/d_c	T/T_c	Calc	Pcnt
0.000039	0.3624	0.3618	0.18
.000066	.3754	.3749	.11
.000108	.3883	.3881	.04
.000171	.4012	.4013	-.01
.000261	.4142	.4144	-.06
.000387	.4271	.4275	-.09
.000561	.4401	.4406	-.12
.000795	.4530	.4536	-.13
.001101	.4660	.4666	-.13
.001496	.4789	.4795	-.12
.001997	.4918	.4924	-.11
.002621	.5048	.5052	-.08
.003390	.5177	.5180	-.06
.004323	.5307	.5308	-.03
.005443	.5436	.5436	-.00
.006774	.5566	.5564	.02
.008340	.5695	.5692	.05
.010166	.5824	.5820	.07
.012280	.5954	.5949	.09
.014709	.6083	.6077	.10
.017483	.6213	.6206	.11
.020632	.6342	.6335	.11
.024188	.6472	.6465	.11
.028186	.6601	.6595	.10
.032664	.6731	.6725	.09
.037658	.6860	.6855	.07
.043211	.6989	.6986	.05
.049369	.7119	.7116	.03
.056178	.7248	.7247	.01
.063697	.7378	.7378	-.01
.071985	.7507	.7509	-.03
.081109	.7637	.7640	-.04
.091132	.7766	.7770	-.06
.102160	.7895	.7901	-.07
.114275	.8025	.8031	-.07
.127589	.8154	.8160	-.07
.142234	.8284	.8289	-.07
.158365	.8413	.8418	-.06
.176176	.8543	.8546	-.04
.195880	.8672	.8674	-.02
.217781	.8801	.8802	-.00
.242234	.8931	.8929	.02
.269734	.9060	.9057	.04
.300976	.9190	.9185	.06
.336923	.9319	.9313	.06
.388905	.9474	.9470	.04
.418343	.9546	.9546	.01
.477367	.9673	.9674	-.01
.509467	.9732	.9732	.01
.627071	.9882	.9884	-.02
.718195	.9950	.9950	-.01

TABLE 5, PART II. Oxygen liquid densities

d/d_c	T/T_c	Calc	Pcnt
1.4126	0.9877	0.9875	0.03
1.4763	.9815	.9813	.02
1.5358	.9742	.9742	-.00
1.6050	.9644	.9643	.01
1.6654	.9542	.9541	.02
1.7188	.9440	.9438	.02
1.7837	.9299	.9298	.01
1.8180	.9218	.9217	.01
1.8578	.9119	.9118	.02
1.8881	.9037	.9037	-.00
1.9104	.8976	.8975	.01
1.9506	.8859	.8859	.00
1.9957	.8718	.8720	-.02
2.0192	.8643	.8644	-.01
2.0502	.8541	.8540	.01
2.0680	.8476	.8479	-.03
2.0914	.8396	.8396	-.01
2.1217	.8284	.8285	-.02
2.1558	.8154	.8156	-.02
2.1890	.8025	.8025	-.00
2.2205	.7895	.7896	-.01
2.2516	.7766	.7766	.01
2.2814	.7637	.7636	.01
2.3099	.7507	.7509	-.02
2.3385	.7378	.7378	-.00
2.3661	.7248	.7248	-.00
2.3936	.7119	.7116	.03
2.4195	.6989	.6990	-.01
2.4459	.6860	.6858	.03
2.4712	.6731	.6730	.01
2.4963	.6601	.6601	.01
2.5209	.6472	.6471	.00
2.5453	.6342	.6342	.00
2.5691	.6213	.6214	-.01
2.5934	.6083	.6081	.04
2.6163	.5954	.5954	-.00
2.6397	.5824	.5823	.02
2.6625	.5695	.5694	.01
2.6848	.5566	.5567	-.02
2.7073	.5436	.5437	-.01
2.7293	.5307	.5308	-.03
2.7516	.5177	.5177	.01
2.7733	.5048	.5048	-.00
2.7943	.4918	.4922	-.07
2.8166	.4789	.4787	.04
2.8371	.4660	.4662	-.05
2.8591	.4530	.4527	.06
2.8791	.4401	.4403	-.06
2.9006	.4271	.4270	.03
2.9212	.4142	.4141	.01
2.9421	.4012	.4010	.07
2.9621	.3883	.3883	-.01
2.9825	.3754	.3755	-.03
3.0030	.3624	.3624	-.01

TABLE 6. Constants and coefficients for equation (5)

	Hydrogen	Oxygen	Fluorine	Neon
T_c , K	32.953	154.52	143.9	44.40
ρ_c , mol/l	15.59	13.52	15.1	23.3
A_1	0.300 6242	0.185 6314	0.180 5343	0.208 9927
A_2	-0.567 0332	-0.323 1999	-0.327 0052	-0.364 1367
A_3	3.838 7436	1.997 6801	2.253 8388	2.785 5198
A_4	-19.378 2878	-5.607 8688	-10.371 1663	-10.531 0711
A_5	89.909 2005	14.689 1100	37.918 2595	27.607 2367
A_6	-237.846 2970	-33.547 8549	-85.040 7456	-49.451 6169
A_7	331.544 2639	48.360 0365	108.079 9417	56.381 3846
A_8	-231.236 7070	-36.011 9980	-71.747 6081	-35.749 6220
A_9	64.190 7466	10.667 7429	19.470 3943	9.500 5538
NP	62	105	58	31
RMS, %	0.0078	0.054	0.0086	0.126

4. Discussion

TABLE 7. Comparisons of critical constants

T_c , K	ρ_c , mol/l	Reference
Parahydrogen		
32.984	15.27 ± 0.30	[9]
32.976	15.59 ± 0.05	[8]
32.934 ^a	15.59 ± 0.02^b	See footnotes.
32.953	15.59	Eq (5), this report.
Oxygen		
.....	13.44	[11]
154.77	12.	[12]
154.565	12.75	[13]
.....	13.333	[14]
.....	13.62	[4]
154.575 ± 0.01	13.628 ± 0.016	[15]
154.54	13.614	Table 1, this report.
154.52	13.52	Eq (5), this report.
Fluorine		
144.	[18]
.....	12.4	[19]
.....	15.0	[20]
143.9	15.1	Eq (5), this report.
Neon		
44.39	23.96	[21]
44.43	[24]
44.40 ± 0.02	[25]
44.40	23.3	Eq (5), this report.

^a L. A. Weber (unpublished) via data of [10].

^b Via rectilinear diameter (R.D.G.).

A long-standing problem for the computation of thermodynamic properties has been the lack of a satisfactory description of the orthobaric densities, or conversely of the saturation temperatures. The extraordinary accuracy of representation seen for H_2 and O_2 in tables 3 and 5, and for F_2 in the rms deviation of table 6, shows that eq (5) provides a solution for this problem. For oxygen in particular we see that (5) is valid over an enormous range of vapor densities. Uncertainties up to one percent in the lowest vapor density data are assigned by Weber [4].

Critical constants estimated via (5) depend upon the form of (5); upon the (often unknown) accuracy of data near T_c ; and upon elimination of points near T_c . With densities derived from PVT data and a vapor pressure equation as in this report, it would be difficult to assign uncertainties to the estimated critical constants.

This report demonstrates the attractive property of eq (5) that values of T_c and ρ_c adequate for many applications are obtained in one step from the complete set of liquid and vapor densities, without the need for data close to the critical density.

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